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June 12, 2013

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Actinide (U-Th-Pa) concentrations and isotopic disequilibrium in surface soils and glassy fallout beads from historical nuclear tests

Abstract

Fallout from a nuclear event contains a mixture of fuel, fission products and environmental contributions. The physical processes controlling the creation of nuclear fallout are poorly understood. In particular, the magnitude of and mechanisms for environmental contributions to near-surface fallout formation are poorly constrained. Here we present actinide (U, Th, and Pa) concentration and isotopic composition data for twenty-eight pieces of glassy fallout along with surface soil samples associated with a historical above-ground uranium-fueled nuclear test. Surface soil samples were collected proximal to the site of the test, yet distal to fallout plumes, in order to characterize the mineralogical and chemical composition of pre-test soil and geology. Soil analysis results demonstrate that surface chemical heterogeneity precludes assumption of a homogeneous soil end-member. Although a natural uranium isotopic composition ($^{235}U/^{238}U = 7.2527 \times 10^{-3}$) can be assumed for the environmental soils, surface soils have a heterogeneous thorium isotopic composition (average $^{230}Th/^{232}Th = 2.8 \times 10^{-6}$).

Glassy fallout measured in this study has higher uranium concentrations than observed in soils by about an order of magnitude, and variable, non-natural uranium isotopic compositions ($^{235}U/^{238}U = 2.321$ to 7.725). Fallout ^{234}U and ^{235}U concentrations are over two orders of magnitude higher than in soils. In contrast, thorium concentrations in fallout are comparable to soils, but are enriched in ^{230}Th relative to soils by about a factor of two. The uranium and thorium isotopic compositions of fallout can be generally interpreted through a two component mixing model, with device nuclear fuel and surface soils as end-members. The data obtained for this study can be used to calculate ^{234}U-^{230}Th and ^{235}U-^{231}Pa model ages that vary from 129 to 314 years before present and 119 to 151 years before present, respectively. Both chronometers result in systematically inaccurate model ages that are tens to hundreds of years older than dates of nuclear fuel production and United States above-ground nuclear weapons testing. Furthermore, model ages do not agree in cases where both chronometers were measured on a single glassy fallout sample. These results, while consistent with a two-component mixing model, preclude a simple interpretation of model ages from fallout.

Introduction

Nuclear fallout is a byproduct of above-ground nuclear tests in cases where the fireball interacts with the surface of the Earth. Formed in the heat of the fireball, fallout particles are, essentially, parcels of surface soils that melt, incorporate nuclear fuel (as well as fission and activation products in low concentrations), and cool rapidly (Glasstone and Dolan, 1977). Fallout particles are easily identified in the field on the basis of glassy matrix, spheroidal to aerodynamic shape, and radioactivity above background levels (Adams et al., 1960). Examination of the chemical and isotopic composition of fallout particles can provide useful attribution information in a post-detonation nuclear forensic investigation. Available data on fallout composition in the open literature are limited, although a few studies have examined the neutron activation and fission product (e.g. Parekh et al., 2006; Belloni et al., 2011; Bellucci et al., 2012) and major and trace element composition (e.g. Eby et al., 2010; Bellucci and Simonetti, 2012) of trinitite (glass from the Trinity event). In this study, we study the utility of the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers to post-detonation nuclear forensic investigation.

The ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers, well-established in geochemistry for determining the ages of rocks and minerals, have been applied to analysis of nuclear forensics samples, such as highly-enriched uranium in nuclear fuel rods (Lamont and Hall, 2005; Varga and Surányi, 2007; Williams and Gaffney, 2011; Eppich et al., 2013) to determine

processing ages, or time of last separation, etc. Here we explore expansion of these techniques to post-detonation materials, in particular, studying the utility of the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers in the age-dating of macro-scale glassy fallout. In this context, it is imperative to specify what exactly is meant by the term "model age". In nuclear forensic investigations of fuel-cycle nuclear materials, the model age of a sample is the age of the most recent uranium or plutonium purification event in the nuclear fuel production process. In ideal cases, model ages calculated using the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers can have analytical uncertainties of less than a year. Inherent in the calculation of model age are two assumptions:

1. Concentrations of the daughter nuclides (^{230}Th and ^{231}Pa, in these systems) following the final purification event are negligible (initial ^{230}Th and ^{231}Pa concentration = 0).

2. No parent-daughter loss and/or gain occurred between the time of final purification and the time of analysis.

If these assumptions hold true during the formation of fallout, and a post-detonation model age can be accurately and precisely measured, timing information relating to the age and origins of the nuclear materials might be able to be inferred. The assumption of closed-system behavior in post-detonation nuclear forensics samples is *a priori* unlikely. Fallout particles primarily consist of melted and quenched surface soils that contain non-negligible concentrations of ^{230}Th and ^{231}Pa in most geologic settings. In addition, fallout formation processes may cause chemical and isotopic fractionation between parent and daughter nuclides such that the record preserved in fallout does not reflect the relative daughter and parent compositions of the fireball or pre-detonation device. None the less, we pursued this study to better understand the relative environmental contributions to these systems, how they have been modified, and in what context they may be interpreted with respect to times and temperatures of formation.

Samples and methods

Sample collection and dissolution

Six surface soil samples were collected from locations proximal to the ground zero of the test. These soil collections are proxies for the surface soils melted and quenched during the formation of fallout. Care was taken to collect away from the path of the fallout plume to minimize contamination of the surface soils, and soils were inspected to insure exclusion of any macroscopic fallout. The surface soils near the test are rocky, dry, contain little to no organic matter, and consist of a mixture of silicate and carbonate minerals. Soil samples were crushed in an agate mortar and pestle to a grain size of silt to clay. A fraction of each crushed soil was weighed and dissolved in clean Teflon beakers using a mineral acid dissolution procedure routine in geochemistry. Initial sample dissolution was performed using a 2:1 to 3:1 mixture of concentrated HNO_3 to concentrated HF. After a period of >24 hours, the samples transformed to a white sediment, indicating that the Si-O bonds had been broken, and that a fluoride had precipitated from solution. To break the fluoride compounds, 1 mL of concentrated $HClO_4$ was added to each beaker, and the samples were dried down to drive off fluoride. Samples were then dissolved in 1 mL concentrated HCl and dried down to remove excess $HClO_4$ and HF. Finally, the samples were dissolved in 6 mL 3 M HCl to produce stock solutions. At this point, all samples were completely in solution, and care was taken to assess that no precipitate formed. The stock solutions were then split for actinide chemical separation and analysis.

Macroscopic (mm-sized) glassy fallout was likewise collected from areas proximal to the site of the test (within ~600 m of ground zero), along the path of the fallout plume. These glasses, separated from soil grains, were washed using 18.2 MΩ H_2O. Each piece was weighed using a micro-balance, with individual masses varying from 7 to 36 mg. Each piece was then placed in a separate Teflon vial and dissolved, using the same dissolution methods as the soil samples. Care was taken to assess that each sample was completely dissolved and that no precipitate formed in the stock solution. The stock solutions were then split for actinide chemical separation and analysis.

Actinide chemical separation and analysis

Uranium concentrations and isotopic compositions were measured on chemically-purified aliquots of bead and soil stock solutions by isotope dilution mass spectrometry (IDMS) using a calibrated ^{233}U spike, following the procedure outlined by Williams and Gaffney (2011). Thorium concentrations and isotopic compositions were measured on chemically-purified aliquots of fallout glass and soil stock solutions by IDMS using a calibrated ^{229}Th spike, also

following the procedure outlined by Williams and Gaffney (2011). Six fallout samples and one soil sample were chosen for Pa chemical separation and analysis by IDMS, which was performed following a procedure outlined by Eppich et al. (2013), and using ^{233}Pa spike-2 (for fallout samples NT-2-12 A, B, and D) and ^{233}Pa spike-3 (for fallout samples NT-2-12 E, K, and L, and the soil sample) described therein. The spiked and chemically-purified aliquots of U, Th, and Pa were measured by multi-collector inductively-coupled plasma mass spectrometry (MC-ICPMS) at Lawrence Livermore National Laboratory. Uranium was measured using a Nu Plasma MC-ICPMS (bead samples NT-2-12 A through L) and a MicroMass IsoProbe MC-ICPMS (All soil samples and fallout samples NT-2-12 O through AR). Thorium and Pa were measured using a Nu Plasma MC-ICPMS.

Results

Actinide (U, Th, and Pa) concentrations and isotopic compositions for the average of five surface soil samples are presented in Table 1. An additional soil sample measured for this study yielded a ^{235}U-enriched uranium isotopic composition well outside of uncertainty of natural uranium (natural uranium ^{235}U/^{238}U = 7.2527 × 10^{-3}). Micro-scale fallout, present as a contaminant likely originating from the fallout plume, is the probable the source of non-natural uranium in this sample. As the soils measured in this study are to be used as proxies for interpreting the contribution of soil to fallout, this contaminated soil was excluded from further interpretation. The five uncontaminated soils have natural ^{235}U/^{238}U ratios, an average uranium concentration of 3.7 ± 1.6 µg/g, and an average ^{234}U/^{235}U ratio of 0.0076 ± 0.0013 (uncertainties on U and Th soil averages are reported as the 2σ standard deviation of five measurements). Average soil Th concentration is 22 ± 2 µg/g and average ^{230}Th/^{232}Th is 2.79 ± 1.5 × 10^{-6}. The single soil sample measured for Pa has a concentration of 0.800 ± 0.012 pg/g. Fallout actinide concentrations and isotopic compositions are presented in Table 1. The fallout glasses vary in uranium concentration from 14.73 to 32.85 µg/g, in ^{235}U/^{238}U from 2.321 to 7.725, and in ^{234}U/^{235}U from 0.01078 to 0.01093. Fallout Th concentrations vary from 18.87 to 26.80 µg/g, and ^{230}Th/^{232}Th ratios vary from 3.659 × 10^{-6} to 4.888 × 10^{-6}. Protactinium concentrations in the fallout vary from 1.82 to 2.45 pg/g.

Discussion

Uranium concentrations in glassy fallout samples are about an order of magnitude higher than those observed in associated surface soil. Less surprisingly, uranium isotope ratios in fallout samples are distinctly different from the natural uranium isotopic ratios of the surface soils. The non-natural, ^{235}U-enriched uranium isotope ratios observed in fallout are consistent with remnant nuclear fuel in the device that did not undergo fission. Fallout samples also have higher ^{234}U/^{238}U ratios than soil samples, with concentrations of ^{234}U and ^{235}U in fallout over two orders of magnitude higher than observed in the soil. Assuming negligible mass loss and/or gain during fallout formation, one would infer that >99 % of the ^{234}U and ^{235}U in the fallout must originate from remnant device fuel.

Fallout and soil Th concentrations are roughly similar. The ^{230}Th concentration in fallout, however, is higher than that observed in soils by about a factor of two. As ^{230}Th is predicted to be present in device nuclear fuel due to decay of ^{234}U, the measured ^{230}Th concentration of fallout samples must result from addition of device ^{230}Th to naturally-occurring Th in the surface soils. Protactinium behaves similarly to ^{230}Th, as ^{231}Pa is predicted to be present in the device due to ^{235}U decay. Enhanced fallout ^{231}Pa concentrations, like ^{230}Th concentrations, must reflect the addition of device ^{231}Pa to naturally-occurring Pa in the surface soils.

The uranium and thorium isotopic composition observed in the fallout samples can be explained using a simple two-component mixing model, using the uncontaminated surface soil and device nuclear fuel as end-members. We can assume highly-enriched uranium as the weapon fuel composition, we assume a device ^{235}U/^{238}U end-member value of 13 (~93 % enriched in ^{235}U). We also assume that the only Th contributed by the device end-member is ^{230}Th (^{232}Th/^{230}Th = 0). The isotopic composition of the soils is used to represent the (nominally) uncontaminated environmental soil end-member. The uranium isotopic composition of our measured soils is within uncertainty of natural uranium (^{235}U/^{238}U = 0.00727 ± 0.00007), and can be considered invariant. The thorium isotopic composition of these soils varies by about a factor of two, however, and the five samples measured for this study may not represent the complete ^{232}Th/^{230}Th range of soils incorporated into fallout. On the basis of our soil measurements, we

assume a range of soil end-member ^{232}Th/^{230}Th, from 2.9×10^5 to 5.3×10^5. All but one of the fallout samples measured in this study fall within the range of isotopic compositions predicted by the two-component mixing model (Figure 1). One fallout sample has a lower ^{232}Th/^{230}Th than predicted by the model, which may be due to the incorporation of surface soils with lower ^{232}Th/^{230}Th values than measured by this study.

This two-component mixing model does not take into account the possibility of chemical and/or isotopic fractionation during the formation of glassy fallout beads, nor does it consider the possibility of mass loss and/or gain. Despite these gross oversimplifications, this model indicates that simple two-component mixing can broadly explain the observed concentrations and isotopic compositions of the actinides in this fallout, suggesting that fractionation and mass loss/gain are minor or negligible processes. Though outside the scope of the present paper, this model can be tested using other elements expected to be present in the device at concentrations significantly higher than surface soils, such as other actinides present in nuclear fuel or non-nuclear device components.

While it is possible to calculate a model age of the nuclear fuel component of fallout using the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers, post-detonation chronology is a more complicated case because the actinide budget of fallout is indicative of two-component mixing between nuclear fuel and soil end-members. The model age (the age of the most recent chemical purification event) can be determined on the basis of the radioactive decay of ^{234}U to ^{230}Th and ^{235}U to ^{231}Pa at a constant rate. Accurate calculation of the model age is only possible if initial daughter nuclide concentrations are negligible (or are known and can be corrected for) and if the parent-daughter pairs behave as closed systems. Although the assumption of complete daughter removal during fuel production may or may not apply (see Williams and Gaffney, 2011 and Eppich et al., 2013 for a thorough treatment of the issue of non-negligible initial daughter nuclide concentrations in the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers, respectively), the assumption of closed-system behavior clearly does not apply in the case of fallout formation. Therefore, model ages calculated using either chronometer are expected to be systematically inaccurate. Incorporation of surface soils with non-negligible concentrations of ^{230}Th and ^{231}Pa should result in model ages older than known dates of nuclear fuel production.

Model ages and associated uncertainties are calculated for bead samples using the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers following Williams and Gaffney (2011) and Eppich et al. (2013), respectively. The results of these calculations are presented in Table 2. Fallout bead model ages calculated using the ^{234}U-^{230}Th chronometer vary from 129 to 314 years before present. For the six beads analyzed for Pa, ^{235}U-^{231}Pa model ages vary from 119 to 151 years before present. ^{234}U-^{230}Th and ^{235}U-^{231}Pa model ages do not agree for bead samples in which both chronometers were used. It is clear that these model ages are inaccurate and biased towards older ages. Accurate model ages would fall within known ages of United States nuclear fuel production and above-ground nuclear weapons testing, which occurred between about 1945 to 1963 (49 to 67 years before these analyses were performed, in 2012).

Theoretically, if the thorium isotopic composition of the surface soil end-member was constant, it would be possible to calculate the nuclear fuel ^{230}Th/^{232}Th, and possible to calculate a ^{234}U-^{230}Th model age (albeit with large uncertainty that may preclude its utility for attribution). This theoretical calculation cannot be performed for the ^{235}U-^{231}Pa chronometer, as Pa is monoisotopic (unless environmental Pa is low enough to be negligible). Due to these limitations, the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers cannot yet be utilized to perform accurate age-dating of fallout particles produced from silicate soils in a post-detonation nuclear forensic investigation. It is possible that the use of these chronometers may be more effective in a geologic environment where surface soil ^{230}Th and ^{231}Pa concentrations are negligible. Fallout produced by historical nuclear weapons tests performed in carbonate shelf environments may be ideal samples to further evaluate the use of these chronometers for nuclear forensic investigations.

Conclusions

1) Glassy fallout beads measured in this study, produced during an above-ground nuclear test of a uranium device, have a non-natural uranium isotopic composition (^{235}U/^{238}U = 2.321 to 7.725). In addition to the naturally-occurring U, Th, and Pa in surface soils, the beads incorporate residual device uranium that did not undergo nuclear fission during the test. Furthermore, the beads incorporate ^{230}Th and ^{231}Pa present in the nuclear fuel due to decay of ^{234}U and ^{235}U, respectively.

2) The uranium and thorium isotopic composition of glassy fallout beads can be broadly explained using a two-component mixing model between surface soil ($^{235}U/^{238}U$ = 0.00727, $^{232}Th/^{230}Th$ = 2.9 × 10^5 to 5.3 × 10^5) and highly-enriched uranium and ^{230}Th derived from the device ($^{235}U/^{238}U$ ≥ 9, $^{232}Th/^{230}Th$ = 0). Observed spread in the data is primarily due to naturally-occurring variability in the $^{232}Th/^{230}Th$ of surface soil incorporated into individual fallout beads.

3) Model ages calculated using the ^{234}U-^{230}Th and ^{235}U-^{231}Pa chronometers are systematically inaccurate, as they are tens to hundreds of years older than dates of United States nuclear fuel production and above-ground nuclear weapons testing. Contamination of nuclear fuel U, Th, and Pa with naturally-occurring U, Th, and Pa in the soil precludes a simple determination of ^{234}U-^{230}Th and ^{235}U-^{231}Pa age-dates. The bias towards older model ages is due to incorporation of soil ^{230}Th and ^{231}Pa in fallout at concentrations comparable to that contributed by the device, relative to the concentration of ^{234}U and ^{235}U, which have a device origin of >99 %. The assumption of closed-system parent-daughter behavior in nuclear fuel is violated due primarily to the acquisition of naturally-occurring ^{230}Th and ^{231}Pa during fallout formation.

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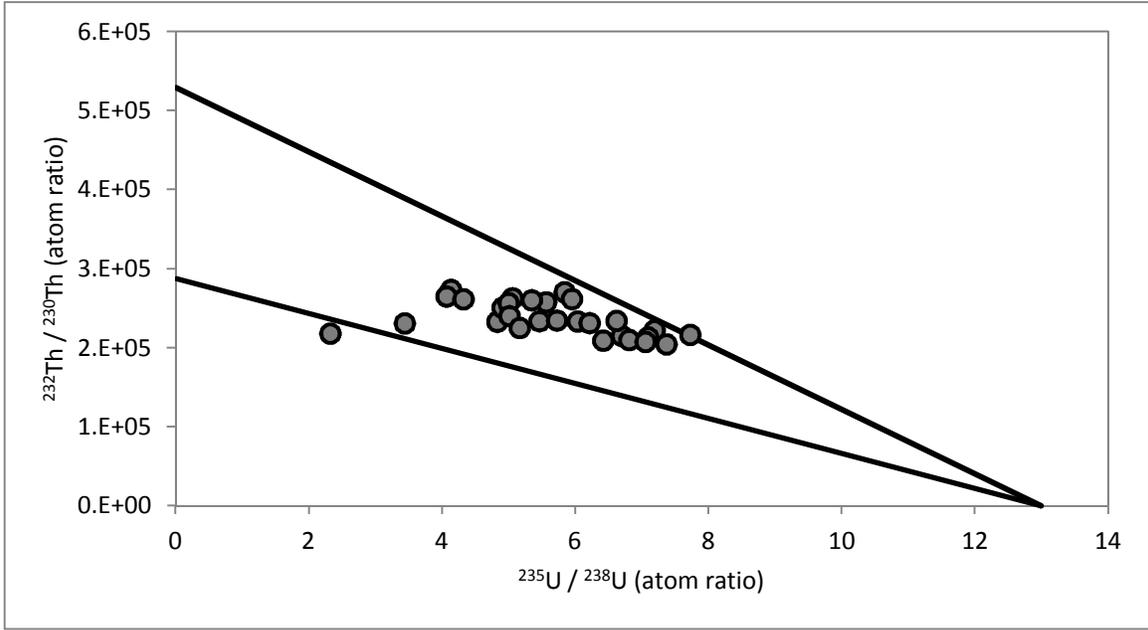
Figure 1. Uranium and thorium isotopic composition of glassy fallout beads. Black lines represent the upper and lower bounds of two-component mixing between a hypothetical nuclear weapon with $^{235}U/^{238}U = 13$ and $^{232}Th/^{230}Th = 0$, and surface soil with $^{235}U/^{238}U = 0.00727 \pm 0.00007$ and $^{232}Th/^{230}Th$ between 2.9×10^5 and 5.3×10^5. Uncertainty bars are smaller than symbols.
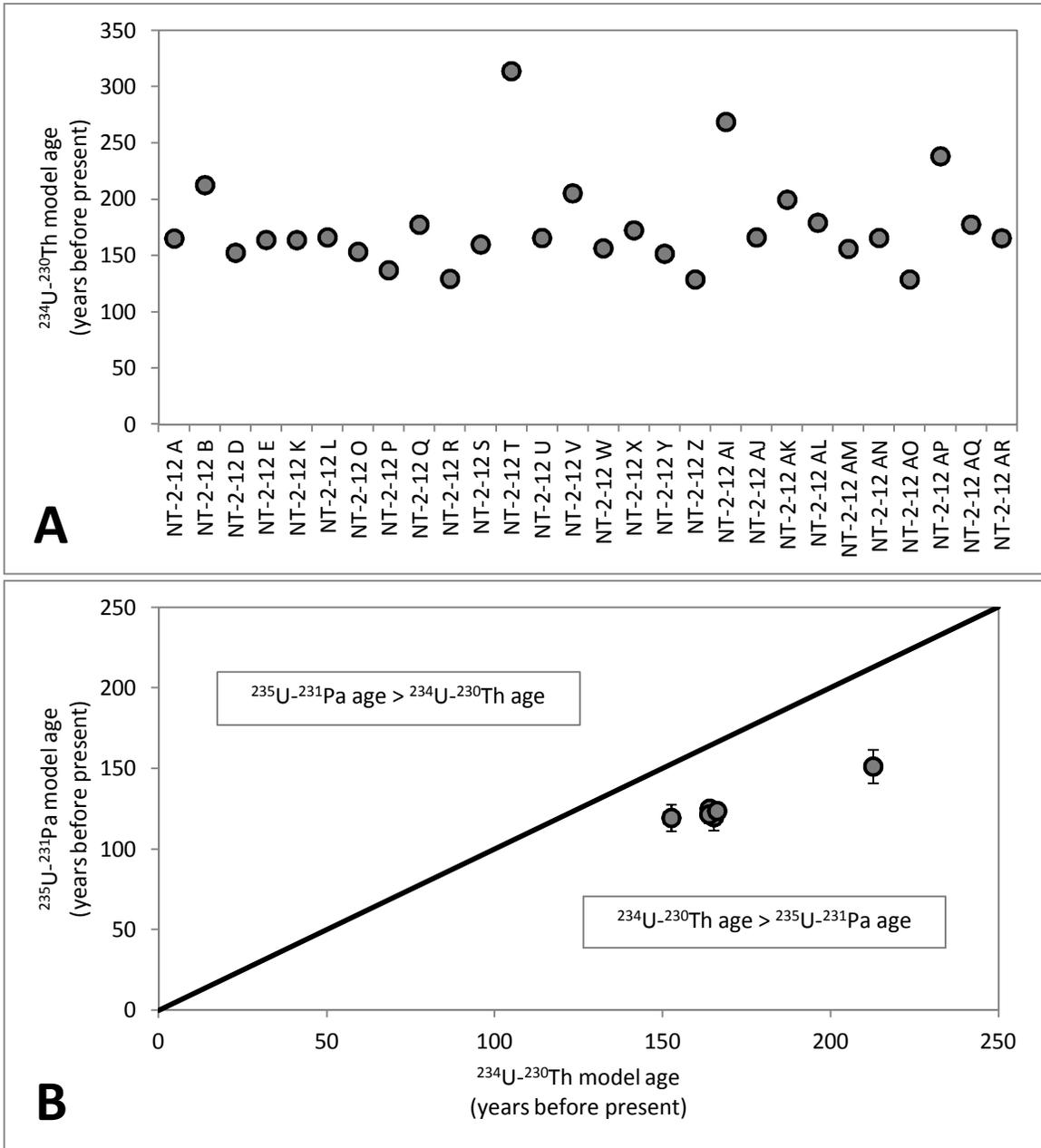
Figure 2. Panel A, ^{234}U-^{230}Th model ages of all glassy fallout beads measured in this study; Panel B, ^{234}U-^{230}Th and ^{235}U-^{231}Pa model ages of glassy fallout beads NT-2-12 A through L. Black diagonal line represents equal ^{234}U-^{230}Th and ^{235}U-^{231}Pa model ages. Uncertainty bars of ^{234}U-^{230}Th model ages are smaller than symbols in both panels.

Sample ID	Mass	U	st. uncert.	$^{235}U / ^{238}U$	st. uncert.	$^{234}U / ^{238}U$	st. uncert.	Th	st. uncert.	$^{230}Th / ^{232}Th$	st. uncert.	Pa	st. uncert.
	(g)	(ug / g)		(atom ratio)		(atom ratio)		(ug / g)		(atom ratio)		(pg / g)	
NT-2-12 A	0.035600	21.400	0.015	5.561	0.004	0.05997	0.00005	22.78	0.07	3.880E-06	1.9E-08	2.06	0.07
NT-2-12 B	0.030618	18.970	0.013	4.134	0.003	0.04459	0.00004	26.14	0.08	3.659E-06	1.8E-08	2.19	0.08
NT-2-12 D	0.024565	19.182	0.014	5.344	0.004	0.05764	0.00005	18.87	0.06	3.843E-06	1.9E-08	1.82	0.06
NT-2-12 E	0.027148	22.102	0.016	5.839	0.004	0.06302	0.00005	24.78	0.07	3.698E-06	1.8E-08	2.25	0.04
NT-2-12 K	0.024625	21.940	0.016	5.057	0.004	0.05456	0.00004	23.36	0.07	3.805E-06	1.9E-08	2.12	0.05
NT-2-12 L	0.024938	25.171	0.018	4.828	0.004	0.05210	0.00004	23.93	0.07	4.292E-06	2.2E-08	2.45	0.05
NT-2-12 O	0.021116	25.64	0.04	5.461	0.006	0.0592	0.0004	23.15	0.06	4.278E-06	1.7E-08	NM	-
NT-2-12 P	0.017437	31.59	0.06	7.189	0.010	0.0779	0.0006	25.11	0.07	4.505E-06	1.8E-08	NM	-
NT-2-12 Q	0.020327	21.16	0.04	4.907	0.007	0.0532	0.0004	23.31	0.06	3.989E-06	1.6E-08	NM	-
NT-2-12 R	0.017022	30.81	0.05	7.093	0.010	0.0770	0.0006	22.22	0.06	4.692E-06	1.8E-08	NM	-
NT-2-12 S	0.015143	24.68	0.04	5.724	0.007	0.0623	0.0005	23.54	0.06	4.263E-06	1.8E-08	NM	-
NT-2-12 T	0.018902	14.73	0.02	2.321	0.003	0.0253	0.0002	21.09	0.05	4.584E-06	2.0E-08	NM	-
NT-2-12 U	0.023776	22.01	0.04	4.998	0.007	0.0544	0.0004	23.30	0.06	3.898E-06	1.6E-08	NM	-
NT-2-12 V	0.019379	17.41	0.03	4.067	0.005	0.0443	0.0003	22.76	0.06	3.773E-06	1.8E-08	NM	-
NT-2-12 W	0.009058	28.00	0.05	6.418	0.008	0.0700	0.0005	23.74	0.06	4.783E-06	2.1E-08	NM	-
NT-2-12 X	0.008650	23.98	0.04	6.698	0.009	0.0732	0.0005	23.23	0.06	4.649E-06	2.7E-08	NM	-
NT-2-12 Y	0.017236	26.47	0.05	6.035	0.007	0.0658	0.0005	24.08	0.06	4.283E-06	1.8E-08	NM	-
NT-2-12 Z	0.018805	31.96	0.06	6.808	0.009	0.0741	0.0006	22.54	0.06	4.765E-06	2.0E-08	NM	-
NT-2-12 AI	0.007086	16.35	0.03	3.439	0.004	0.0371	0.0002	23.31	0.06	4.331E-06	3.2E-08	NM	-
NT-2-12 AJ	0.010145	22.86	0.04	6.214	0.007	0.0670	0.0003	22.42	0.06	4.323E-06	2.4E-08	NM	-
NT-2-12 AK	0.009069	20.19	0.03	5.950	0.006	0.0641	0.0003	26.80	0.07	3.820E-06	2.0E-08	NM	-
NT-2-12 AL	0.011855	22.70	0.04	5.010	0.005	0.0540	0.0003	24.14	0.06	4.163E-06	1.8E-08	NM	-
NT-2-12 AM	0.007955	31.45	0.05	7.369	0.008	0.0794	0.0004	26.17	0.07	4.888E-06	2.2E-08	NM	-
NT-2-12 AN	0.007408	25.39	0.04	7.053	0.007	0.0760	0.0004	22.64	0.06	4.816E-06	2.9E-08	NM	-
NT-2-12 AO	0.012177	32.85	0.06	7.725	0.008	0.0833	0.0004	24.02	0.06	4.622E-06	2.3E-08	NM	-
NT-2-12 AP	0.007856	16.17	0.03	4.320	0.004	0.0466	0.0002	24.25	0.06	3.824E-06	2.5E-08	NM	-
NT-2-12 AQ	0.010257	23.55	0.04	5.165	0.005	0.0557	0.0003	23.36	0.06	4.442E-06	2.2E-08	NM	-
NT-2-12 AR	0.011988	22.90	0.04	6.619	0.007	0.0714	0.0003	22.82	0.06	4.272E-06	2.0E-08	NM	-
average soil	-	3.7	1.6	0.00727	0.00007	5.6E-05	9E-06	22	2	2.79E-06	1.5E-06	0.800	0.012

Table 1. Actinide concentrations and isotopic compositions of glassy fallout beads and surface soils. Surface soil data is an average of the measurement of five samples, except for Pa, which is from a single measurement. Uncertainty on the average surface soil is calculated as the 2σ standard deviation of five meausurements, except for Pa. NM, not measured.

Sample ID	^{234}U-^{230}Th model age (years)	st. uncert.	^{235}U-^{231}Pa model age (years)	st. uncert.
NT-2-12 A	165.2	1.3	120	8
NT-2-12 B	212.6	1.8	151	10
NT-2-12 D	152.5	1.3	119	8
NT-2-12 E	163.9	1.3	125	5
NT-2-12 K	163.8	1.4	122	5
NT-2-12 L	166.2	1.5	124	5
NT-2-12 O	153	3	NM	-
NT-2-12 P	137	2	NM	-
NT-2-12 Q	178	3	NM	-
NT-2-12 R	129	2	NM	-
NT-2-12 S	160	3	NM	-
NT-2-12 T	314	6	NM	-
NT-2-12 U	166	3	NM	-
NT-2-12 V	205	4	NM	-
NT-2-12 W	156	3	NM	-
NT-2-12 X	172	3	NM	-
NT-2-12 Y	152	3	NM	-
NT-2-12 Z	129	2	NM	-
NT-2-12 AI	269	5	NM	-
NT-2-12 AJ	166	3	NM	-
NT-2-12 AK	200	3	NM	-
NT-2-12 AL	179	3	NM	-
NT-2-12 AM	156	2	NM	-
NT-2-12 AN	166	3	NM	-
NT-2-12 AO	128.8	2	NM	-
NT-2-12 AP	238	4	NM	-
NT-2-12 AQ	178	3	NM	-
NT-2-12 AR	165	2	NM	-

Table 2. ^{234}U-^{230}Th and ^{235}U-^{231}Pa model age calculation results for glassy fallout beads. Model ages and associated uncertainties were calculated for the ^{234}U-^{230}Th chronometer following Williams and Gaffney (2011) and for the ^{235}U-^{231}Pa chronometer following Eppich et al. (2013). NM, not measured.